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Materials Science Division

May 1976

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ABSTRACT

Radioactive ^{85}Kr is used as a tracer to detect pinhole failures in GCFR cladding. High-purity helium (99.99% pure) that contains 0.3 ppm ^{85}Kr is used to pressurize the tubular test specimens, and a Geiger-Mueller counter is used to detect ^{85}Kr in the helium environmental gas as it leaves the test chamber. Under the least favorable conditions of temperature and specimen pressure (760°C and 35.6 atm), it is estimated that the smallest pinhole failure that could be detected within 60 s would have an orifice diameter of 0.0102 cm (~102 μm). Using lead shielding around the Geiger-Mueller counter to reduce background radiation, the electronics associated with the ^{85}Kr detector will terminate a biaxial creep test at ^{85}Kr activity levels above 20 counts/min.

I. INTRODUCTION

The purpose of the present report is fivefold: (a) to describe the use of radioactive krypton (^{85}Kr) for the detection of pinhole failures in GCFR cladding, (b) to inform interested and affected parties of plans to use ^{85}Kr in Room CL-103 of Building 212 at Argonne National Laboratory, (c) to acquaint all parties of the manner in which the ^{85}Kr will be used and exhausted to the building stack, (d) to define the minimum ^{85}Kr concentration required for the intended application, and (e) to present data in support of the view that the quantities involved and the method of discharge will not exceed ERDA standards for the maximum permissible concentration (mpc) of ^{85}Kr in air. On the basis of the information presented, it is proposed that the quantities of ^{85}Kr to be used will not adversely affect ANL personnel working in the immediate vicinity of Room CL-103 nor be detrimental to the general public.

II. USE OF KRYPTON-85 IN GCFR CLADDING STUDIES

High-purity helium tagged with parts-per-million quantities of ^{85}Kr will be used to internally pressurize tube specimens during the mechanical-property studies of Gas-Cooled Fast Reactor (GCFR) cladding. Short-term burst and long-term biaxial creep tests will be conducted in a helium environment flowing at a volume rate of 400 cc/min. These tests will encompass a temperature range between 538 and 760°C, and the specimens will be subjected to internal pressures between 526 and 7853 psig. The test apparatus was used in similar studies on Liquid Metal Fast Breeder Reactor (LMFBR) cladding and has been described in detail elsewhere.[1] A simplified representation of the overall test system is shown in Fig. 1.

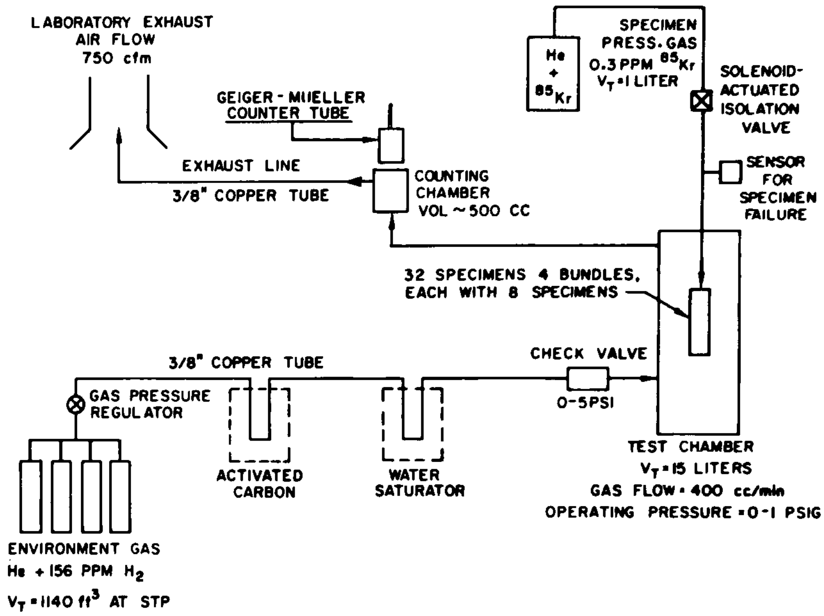

Fig. 1. Schematic of Gas Flow through GCFR Cladding Test Equipment. Neg. No. MSD-62620.

The test apparatus accommodates 32 specimens arranged in four bundles or clusters, each containing eight specimens. The eight specimens of one bundle are attached to a common manifold and are pressurized simultaneously from a 1-liter, high-pressure gas-storage vessel. Each specimen bundle is pressurized from a different gas-storage vessel, and each pressurizing system contains a pressure gauge with electrical contacts. When specimen failure results in a drop in pressure, the closing of the electrical contacts actuates the solenoid valve in the pressurizing line, which automatically isolates the specimen bundle from its high-pressure gas source. When specimen failure by wall perforation is so small that ^{85}Kr can leak out without a drop in specimen pressure, the Geiger-Mueller (G-M) counter in the effluent gas stream will detect the ^{85}Kr activity and also initiate the closing of the solenoid isolation valve. Therefore, the ^{85}Kr tracer released to the test chamber will be exhausted to the building stack through a 3/8-in.-outside-diameter (OD) copper exhaust line.

The environmental gas is commercial high-purity helium that contains a maximum of 156 ppm H_2. It is supplied by four gas bottles, each at 2640 psig pressure, and each contains ~285 ft^3 (8067 liters) of gas at standard temperature and pressure (STP) conditions. The environmental gas contains a maximum of 38 ppm H_2O before it enters the test chamber through a 3/8-in.-OD copper line. A regulated supply-line pressure of 5-20 psig is expected to produce the 400-cc/min gas flow rate through the test chamber, which will operate at or near atmospheric pressure (0-10 in. of H_2O). The check valve in the supply line will prevent back-pressure surges and possible ^{85}Kr flow toward the environmental gas supply.

The 156 ppm of H_2 in the environmental helium gas is well below the 8% H_2 concentration that has been found inherently safe with respect to explosion hazard when mixed with air in all proportions.[2,3] The subsequent addition of 38 ppm H_2O is not expected to adversely affect the inherently safe character of the environmental gas mixture. Argon-H_2 mixtures will be avoided because of the potential explosion hazard identified with certain H_2 concentrations.[4]

The bottles of helium gas that supply the gas environment will be replenished in pairs to avoid unscheduled test interruptions. The depletion rate of gas-bottle pairs at a gas flow rate of 400 cc/min will be

$$(2 \times 8067 \times 2525/2600) \div (0.4 \times 60 \times 24) = 27 \text{ days.}$$

Therefore, the environmental gas bottles will be replaced in pairs at 27-day intervals.

III. KRYPTON-85 CONCENTRATIONS

Test specimens will be pressurized with a helium-^{85}Kr gas mixture that contains 0.3 ppm ^{85}Kr supplied by a standard gas cylinder at 2200 psig pressure. The total gas volume at STP is 213 ft^3 (6029 liters). The 2.7-Ci activity level of the cylinder will be below the 3-Ci/cylinder activity limit imposed by transportation regulations. All quantities of ^{85}Kr released to the test chamber will be exhausted, with the environmental gas, to the atmosphere through the building stack. The rationale and the calculation methods used in this assessment are given in Appendixes A-C and Figs. 2-8 (in Appendix C).

Figure 4 shows the initial or maximum ^{85}Kr flow rates as a function of total pressure for two orifice sizes. These data assume a 0.3-ppm ^{85}Kr concentration in helium and encompass the temperature and pressure ranges of interest, namely, 538-760°C and 523-7853 psig. Pinhole-type cladding failures are simulated by orifice diameters of 2.54×10^{-4} and 2.54×10^{-3} cm. (Hereafter, to facilitate calculations, specimen pressures will be given in atmospheres or dynes/cm^2.*) Data in Fig. 4 indicate that, for a given orifice size (2.54×10^{-3} cm), the greatest difficulty encountered in the detection of pinhole-type specimen failures will be at the lowest specimen pressure and the highest

*1 atm = 14.696 psi = 1.0132×10^6 dynes/cm^2.

specimen temperature. Since cladding failure by violent rupture is often preceded by a pinhole leak, rapid detection of the smallest pinhole leak is important to the study of failure mechanisms.[5] In all the following calculations, rapid and complete mixing is assumed between the ^{85}Kr released from the specimen and the environmental gas in the test chamber.

Figure 5 shows that, at 760°C and a specimen pressure of 35.6 atm, the ^{85}Kr flow rate through an orifice with a diameter of 2.54×10^{-3} cm remains essentially constant for the first 1000 s and then begins to decrease appreciably. Figure 6 indicates that the volume release of helium and ^{85}Kr to the test chamber through a cladding pinhole failure of the same orifice size is also constant during the first 1000 s. Since the flow rate of the environmental gas past the G-M counter located in the effluent gas stream is 400 cc/min, the ^{85}Kr released to the test chamber will be swept past the detector at the same rate.

The capability of the G-M counter to detect a specimen failure of this size depends upon the concentration of ^{85}Kr passing the counter per minute and the counter efficiency. The latter is affected by the estimated 50% areal transparency of the stainless steel window in the counting chamber (5.08×10^{-3} cm in diameter by 7.62×10^{-3} cm thick) and the efficiency with which the counter is shielded against normal background radiation. For a totally unshielded counter, the normal background radiation produces ~200-400 counts/min. The gas-exhaust system is constructed so that 100% of the effluent gas stream passes the G-M counter.

Figure 6 shows that 3.1×10^{-8} liter of ^{85}Kr and 6.5×10^{-2} liter of helium will be released to the test chamber during the first 60 s through a pinhole failure with an orifice diameter of 2.54×10^{-3} cm when a specimen is pressurized to 35.6 atm. Since both these volumes are small compared with the 15-liter test-chamber volume, ^{85}Kr dilution will be determined solely by the volume of the test chamber. The activity of the ^{85}Kr released to the test chamber and the capability of the G-M counter to dectect the activity can be determined by using the decay constant for ^{85}Kr from Appendix B [$\lambda = 1.22791 \times 10^{-7}$ disintegration per minute (dpm)/atom ^{85}Kr].

(a) <u>Activity of ^{85}Kr released to the test chamber in 60 s</u>

$$[^{85}\text{Kr (liters)} \times \lambda \times 6.03 \times 10^{23}] \div 22.4 = \text{dpm}$$

$$(3.1 \times 10^{-8} \times 1.22791 \times 10^{-7} \times 6.03 \times 10^{23}) \div 22.4 = 1.02 \times 10^{8} \text{ dpm.}$$

(b) <u>Concentration of ^{85}Kr in the test chamber in 60 s</u>

$$^{85}\text{Kr activity} \times \text{dilution factor} = \text{dpm/liter}$$

$$(1.02 \times 10^{8}) \times (3.1 \times 10^{-8} \div 15) = 2.11 \times 10^{-1} \text{ dpm/liter.}$$

(c) Activity ^{85}Kr passing G-M counter in 60 s

^{85}Kr concentration x flow rate = dpm

$2.11 \times 10^{-1} \times 0.4 = 8.44 \times 10^{-2}$ dpm.

Considering a 50% areal transparency of the window in the counting chamber, the ^{85}Kr activity detected by the G-M counter will be $0.5 \times 8.44 \times 10^{-2} = 4.22 \times 10^{-2}$ dpm. This level of activity would not be detected by the G-M counter, even when the counter is heavily shielded. (A 2-in.-thick lead shield around the G-M counter is expected to reduce background radiation to ~10 counts/min.) Since a factor-of-10 increase in orifice size produces a 10^4 increase in flow rate (Fig. 4), the diameter of the smallest pinhole failure detected within 60 s under least favorable conditions of temperature and specimen pressure (Figs. 5 and 6) is estimated to be $\sim 1.02 \times 10^{-2}$ cm. The ^{85}Kr activity released by an orifice of this size and detected by the G-M counter is given by $(R_2^4 \div R_1^4) \times 4.22 \times 10^{-2}$ dpm, where R_1 and R_2 are the radii of the orifice. Thus, the ^{85}Kr activity detected by the G-M counter during the first 60 s after the pinhole failure occurs is

$$\frac{1.06 \times 10^{-9}}{2.601 \times 10^{-12}} \times 4.22 \times 10^{-2} = 17.2 \text{ dpm.}$$

The mpc level for ^{85}Kr discharged to the atmosphere is limited to 6.66×10^2 dpm/liter by ERDA regulations.[6] Using a 2.12×10^4 ventilator dilution factor[7] (750-cfm airflow) and a 10^5 stack dilution factor,[8] the ^{85}Kr concentration exhausted to the stack, and subsequently to the atmosphere within the Laboratory perimeter, in the first 60 s will be:

(a) Stack

[17.2 (dpm) ÷ (flow rate)] x ventilator dilution

$(17.2 \div 0.4) \times 2.12 \times 10^{-4} = 9.11 \times 10^{-3}$ dpm/liter.

(b) Atmosphere within Laboratory perimeter

$9.11 \times 10^{-3} \times 1 \times 10^{-5} = 9.11 \times 10^{-8}$ dpm/liter.

These quantities are 1.37×10^{-6} and 1.37×10^{-10} mpc, respectively, and remain essentially constant for 1000 s and then decrease (Figs. 5 and 6). Therefore, the release of these concentrations of ^{85}Kr to the atmosphere are of no consequence to the environment.

IV. EFFLUENT GAS ACTIVITY

The largest ^{85}Kr concentrations exhausted to the atmosphere with the effluent gas stream will occur after simultaneous failure by violent rupture of several specimens in each of the four specimen bundles and by malfunction of the isolation valve in each pressurizing system (Fig. 2). Should these unlikely events occur, the entire gas mixture of high-pressure helium and 0.3 ppm ^{85}Kr in each of the four storage vessels will discharge into the test chamber. The depressurization of a single gas-storage vessel at an initial pressure of 534.36 atm is characterized by the curves in Figs. 7 and 8. The chronology of events during such rapid depressurization is expected to be as follows.

The high flow rate that results from the sudden release of the helium-^{85}Kr gas mixture will cause the check valve in the environmental gas supply line to close and prevent back-streaming of ^{85}Kr to the environmental gas source. Since the test chamber operates essentially at atmospheric pressure, the discharging gas will exhaust continuously to the stack. Assuming each of the four storage vessels are at the maximum pressure of 534.36 atm when the event occurs, the discharges of helium and ^{85}Kr to the stack can be determined by means of Figs. 7 and 8. Figure 7 shows that flow rates for helium and ^{85}Kr will be 5.2×10^3 and 3.2×10^{-3} cc/s, respectively, and will remain relatively constant for the first 60 s of the depressurization. Depressurization is complete in ~420 s (~7 min). Furthermore, the volume of helium discharged from each storage vessel is larger by a factor of 345 than the test chamber volume; therefore, the ^{85}Kr dilution factor is essentially that represented by the gas mixture of helium and 0.3 ppm ^{85}Kr.

The volumes of helium and ^{85}Kr released to the room ventilator duct will be a maximum during the first 60 s of depressurization and then will decrease significantly over the succeeding 3600 s (60 min), as indicated in Fig. 7. Figure 8 shows that the volumes of helium and ^{85}Kr released from each storage vessel to the room-ventilator duct during the first 60 s will be 2.5×10^2 and 5.6×10^{-5} liters, respectively, and the helium dilution factor will be the ratio of these volumes (2.24×10^{-7}).

(a) ^{85}Kr activity at the ventilator duct in 60 s

$(4 \times 5.6 \times 10^{-5} \times 6.03 \times 10^{23} \times 1.22791 \times 10^{-7}) \div 22.4 =$
7.40×10^{11} dpm.

(b) ^{85}Kr concentration at the ventilator duct in 60 s

$(7.40 \times 10^{11} \div 0.4) \times 2.24 \times 10^{-7} \times 2.12 \times 10^{-4} =$
8.78×10^{1} dpm/liter $\simeq 1.3 \times 10^{-1}$ mpc.

(c) ^{85}Kr concentration released to the atmosphere in 60 s

$8.78 \times 10^{1} \times 1 \times 10^{-5} = 8.78 \times 10^{-4}$ dpm/liter $\simeq 1.3 \times 10^{-6}$ mpc.

During the second 60 s of storage-vessel depressurization, ~6×10^2 liters of helium and 1.68×10^{-4} liter of ^{85}Kr will be exhausted to the stack, and the helium dilution factor will be ~2.8×10^{-7}. The ^{85}Kr concentration released to the atmosphere will be 8.06×10^{-4} dpm/liter or 1.21×10^{-6} mpc. For all subsequent 1-min intervals to complete depressurization of all storage vessels, the amount of ^{85}Kr released will diminish; therefore, the atmospheric concentration should never exceed or approach the 6.66×10^2-dpm/liter mpc set by ERDA regulations. Additional precautions will be taken to ensure safe utilization of the radioactive ^{85}Kr gas species and will also include the following:

1. The entire system (Fig. 1) will be checked for helium tightness at pressures up to 15 psig. This will ensure complete containment of the ^{85}Kr within the test apparatus before it is discharged to the stack.

2. No more than two storage vessels will be in operation at any one time at the maximum pressure of 534.36 atm.

3. Equipment operators and other personnel occupying Room CL-103 will be required to carry radiation monitors.

4. Air monitors will be used in Room CL-103 as recommended by the Building 212 radiation-safety representative.

5. All lines will be purged with 100% helium gas before the apparatus is opened at the conclusion of each test run or when a test run is interrupted.

APPENDIX A

Gas Flow through an Orifice

It is assumed that helium and ^{85}Kr flow rates through an orifice at elevated temperature are adequately represented by[9]

$$\text{Flow rate} = \frac{\pi P_d R^4}{8 \ell \eta} \text{ cc/s,} \tag{A.1}$$

where

P_d = pressure difference across orifice, in dynes/cm^2;

R = orifice radius, in centimeters;

ℓ = orifice length, in centimeters;

and

η = gas viscosity, in poises.

Flow-rate calculations derived from Eq. A.1 assume that a circular orifice is an adequate simulation of a pinhole-type specimen failure. Since pinhole failures in LMFBR cladding occur at low diametral strains (~4%),[5] the orifice length (ℓ) is taken as 4.389×10^{-2} cm (~$0.96 \times$ specimen wall).

APPENDIX B

Krypton-85 Decay Constant

Decay of any radioactive atom species is given by

$$N = N_0 \exp(-\lambda t), \qquad \text{(B.1)}$$

where

N_0 = number of radioactive atoms at reference time t;

N = radioactive atoms after time Δt;

and

λ = decay constant, in dpm/atom.

Rewriting Eq. B.1 and setting $N_0/N = 2$ when $t_{1/2} = 10.74$ yr, λ is calculated as

$$\lambda = (\ell n\ 2) \div (10.74 \times 365 \times 24 \times 60),$$

or

$$\lambda = 1.22791 \times 10^{-7} \text{ dpm/atom.}$$

APPENDIX C

System-volume Relations

A simplified representation of one of four specimen-pressurizing systems in the Mark-II Biaxial Creep Tester is shown in Fig. 2. During long-term biaxial creep tests, each of the four gas-storage vessels (V_{GS}) operates at a different pressure ($P_1 > P_2 > P_3 > P_4$) and each pressurizes a separate bundle of eight specimens. Approximately 100 in. of high-pressure tubing connects each specimen bundle to its high-pressure gas source. A solenoid-actuated valve isolates each specimen bundle from its high-pressure gas source when specimen failure has occurred. About 80-85% of the volume of each specimen is displaced with a solid slug to reduce the volume of high-pressure gas within the specimen. This approach eliminates deformation after specimen failure has occurred while the high-pressure gas is being dissipated.

Storage-vessel volume (V_{sv}) 1 liter

Specimen volume (V_s) 3.50 cc

Pressure-line volume ($V_{p\ell}$) 1.24 cc

Displacement-slug volume (V_{ds}) 2.87 cc

$$V_{sys} = V_{sv} + V_{p\ell} + (V_s - V_{ds}),$$

and

$$V_{sv} \approx 0.99\ V_{sys},$$

where V_{sys} is the system volume. Ideal gas behavior is assumed for the helium-^{85}Kr gas mixture, because both gas species have relatively low critical pressures (P_c) and temperatures (T_c).[8] (For helium, P_c = 2.26 atm and T_c = -269.9°C; for ^{85}Kr, P_c = 54.3 atm and T_c = -63.8°C.)

The gas viscosity data in Fig. 3 (Ref. 9) and Eq. A.1 are used to calculate helium and ^{85}Kr flow rates (cc/s) and volumes (liters) released to the test chamber as a result of specimen failure or sudden depressurization following equipment malfunction. Maximum ^{85}Kr flow rates in a gas mixture that contains 0.3 ppm ^{85}Kr are plotted in Fig. 4 as a function of total system pressure. These data encompass the temperature range 538-760°C and the pressure range 35.6-534.36 atm for flow rates through orifices with diameters of 2.54×10^{-4} and 2.54×10^{-3} cm. Figure 4 shows that the ^{85}Kr flow rate (a) increases by a factor of 15 as the system pressure increases from 35.6 to 534.36 atm at constant temperature and orifice size, (b) decreases by a factor of 1.17 as the gas temperature increases from 538 to 760°C at constant system pressure and orifice size, and (c) increases by a factor of 10^4 for a factor-of-10 increase in orifice size, at constant gas temperature and pressure.

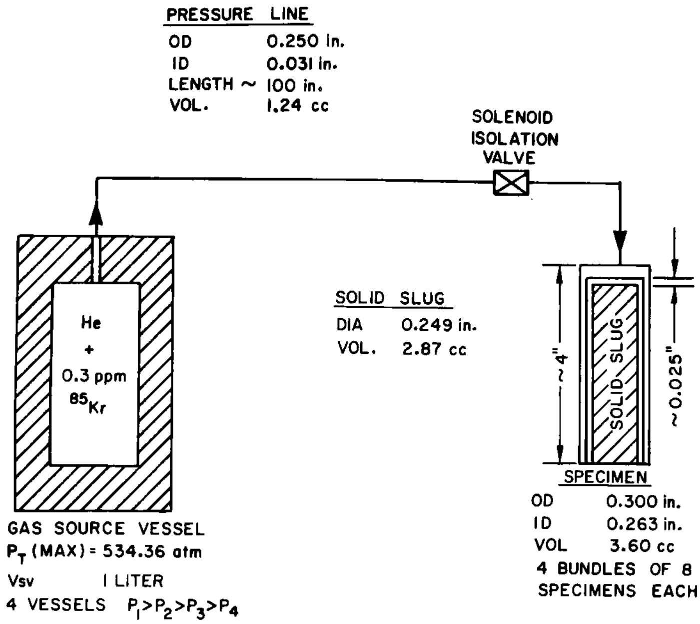

Fig. 2. Volume Relations in GCFR Cladding Test Equipment. Neg. No. MSD-62619.

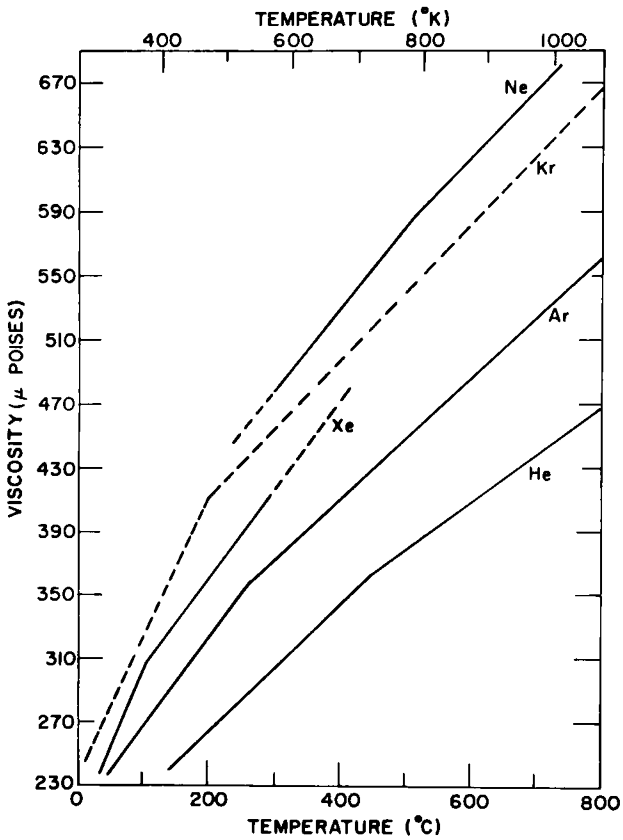

Fig. 3. Viscosities of Noble Gases. Neg. No. MSD-62621.

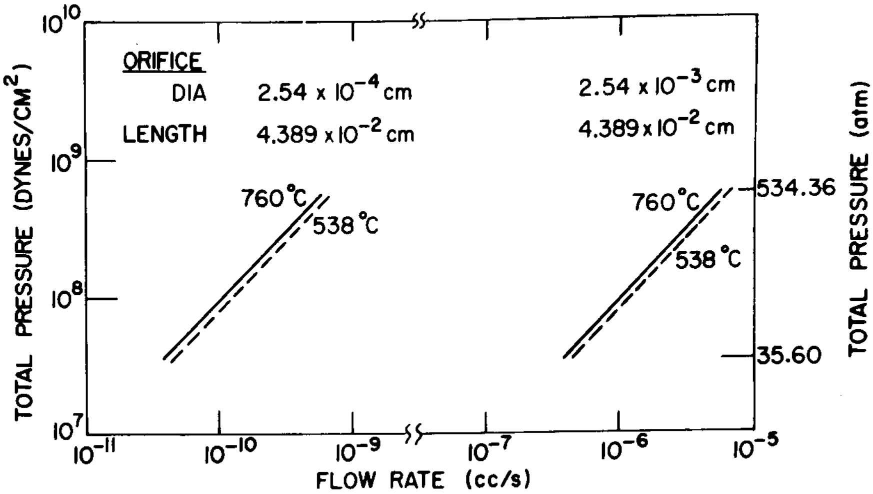

Fig. 4. Maximum ^{85}Kr Flow Rate as a Function of Total Pressure for a Mixture of Helium and 0.3 ppm ^{85}Kr. Neg. No. MSD–62622.

Figures 5 and 6 show the flow rates of helium and ^{85}Kr and the volumes of both gases released as a function of time through a pinhole-type specimen failure simulated by an orifice with a diameter of 2.54×10^{-3} cm. These data are used to assess the capability of the G-M counter to detect pinhole-type specimen failures.

Figures 7 and 8 show the flow rates of helium and ^{85}Kr and the volumes of both gas species released as a function of time during sudden depressurization following equipment malfunction. These data are used to assess the maximum level of ^{85}Kr activity released to the atmosphere through the Building 212 stack.

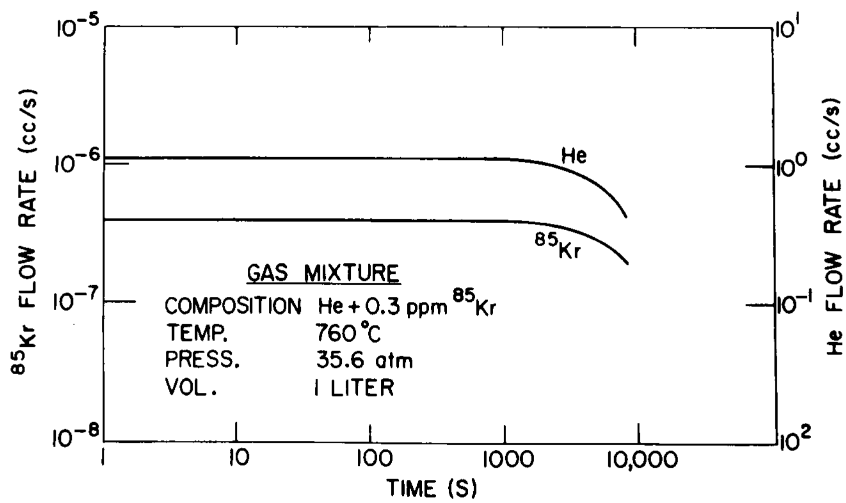

Fig. 5. Flow Rates of Helium and ^{85}Kr for a Gas Mixture Discharging through an Orifice with a Diameter of 2.54×10^{-3} cm. Neg. No. MSD–62623.

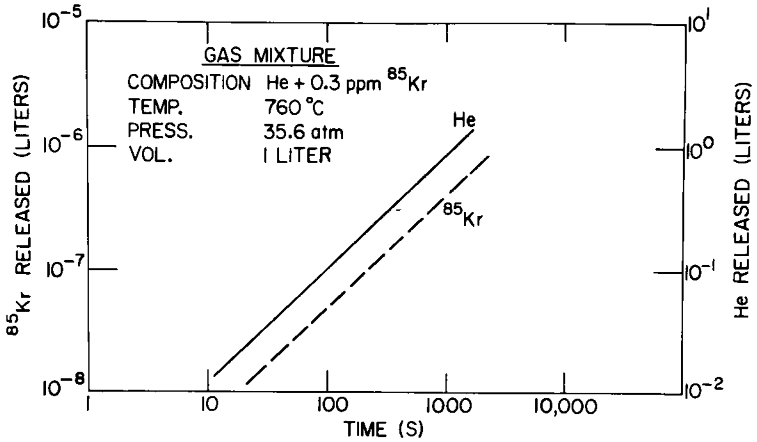

Fig. 6

Volumes of Helium and ^{85}Kr Released by a Gas Mixture Discharging through an Orifice with a Diameter of 2.54 x 10^{-3} cm. Neg. No. MSD-62624.

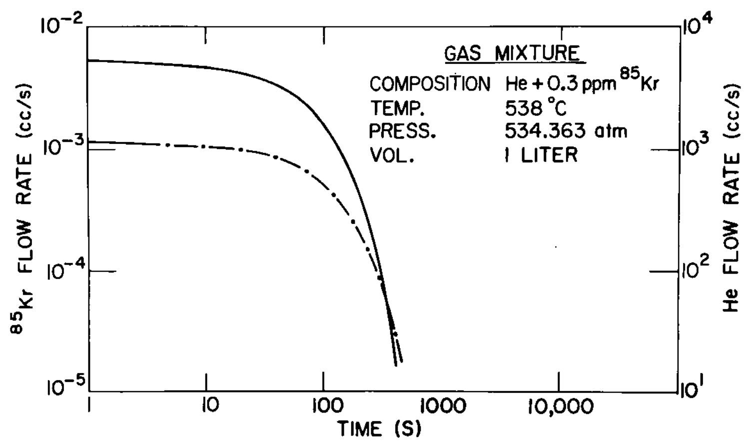

Fig. 7

Flow Rates of Helium and ^{85}Kr for a Gas Mixture Discharging through an Orifice with a Diameter of 7.87 x 10^{-2} cm. Neg. No. MSD-62626.

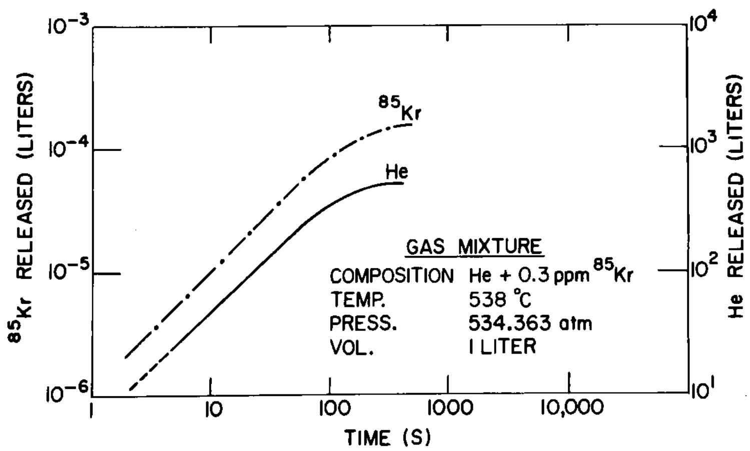

Fig. 8

Volumes of Helium and ^{85}Kr Released by a Gas Mixture Discharging through an Orifice with a Diameter of 7.87 x 10^{-2} cm. Neg. No. MSD-62625.

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